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ROY WALDO MINER

THE SPIN OF THORIUM C′

BY

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ARTHUR BEISER*

Introduction

In 1911, Geiger and Nuttall[1] discovered that the decay constants λ of alpha emitters in the same radioactive series (*i.e.* uranium-radium, actinium, thorium) increase exponentially with alpha particle energy. Plotting log λ versus energy therefore yields a straight line joining the observed values. An apparent exception to the Geiger-Nuttall relation is the decay of ThC′ (${}_{84}\text{Po}^{212}$), which is located significantly below the line joining the other members of the thorium decay series. This is evident from FIGURE 1. No explanation for the deviation is given in the literature, other than the assumption of unreliable experimental data. Both λ and E for this decay, however, have been redetermined with a high degree of accuracy on several occasions[2] without obtaining any better agreement with the Geiger-Nuttall relation.

One method of approaching the anomalous status of ThC′ is to inquire whether any factors might exist that would tend to increase its half life against decay, thus decreasing λ. Such a factor would be the emission of an alpha particle with an angular momentum l greater than zero. In that case the "centrifugal" potential $h^2 l(l+1)/2\mu r^2$ must be added to the Coulomb potential $2Ze^2/r$ used in calculating the lifetime by the Gamow theory.[3] The higher potential then leads to an increased lifetime.

A value of $l > 0$ for the ThC′ → ThD alpha particle requires that, since ThD (${}_{82}\text{Pb}^{208}$) has spin $I = 0$,[4] ThC′ have $I = l$. But ThC′ is an even-even nucleus and, without exception, the known ground states of stable even-even nuclei have $I = 0$. This empirical rule is usually generalized to include unstable even-even nuclei, which are ordinarily not susceptible to measurement, as well. Hence one cannot simply assume that $I > 0$ for ThC′. The experiment described below was designed to determine the spin of ThC′ directly in order to clarify this situation.

(2) *Theory*

ThC′ is a particularly fortunate nucleus to study, since its half-life is only 3×10^{-7} sec. This leads to the possibility that an angular

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correlation might be present in the successive beta and alpha decays in the series

$$\underset{(\mathrm{ThC})}{{}_{83}\mathrm{Bi}^{212}} \xrightarrow{\beta} \underset{(\mathrm{ThC}')}{{}_{84}\mathrm{Po}^{212}} \xrightarrow{\alpha} \underset{(\mathrm{ThD})}{{}_{82}\mathrm{Pb}^{208}} \qquad (1)$$

For a decay chain of this kind, a characteristic distribution in the angle between the directions of motion of the emitted electron and alpha particle may be expected if all of the following conditions are satisfied:[5,6] (1) the spin of the intermediate nucleus is different from zero; (2) the beta transition is forbidden and its energy spectrum has a forbidden shape; and (3) the angular momentum carried off by the alpha particle is greater than zero, and the decay is sufficiently rapid to prevent any perturbation of the intermediate state.

Here the second condition is fulfilled, since the beta-decay of ThC is first-forbidden (log $ft = 7.2$). Recent work[7] indicates that its Kurie plot exhibits the characteristic upward convexity of forbidden spectra in the high-energy region. The latter effect is somewhat decreased here because of the existence of another beta group of lower energy from ThC (leading to an excited state in ThC′) whose end point is near the convexity.

In the limit of zero electron momentum, any correlation must disappear;[5,6] hence those electrons with high energies should show a stronger correlation with the alpha particle than the less energetic ones. Low-energy betas can be experimentally discriminated against, however, and so this factor presents no difficulty.

The last part of condition 3 is presumably satisfied by the short lifetime of ThC′. It has already been established[4] that ThD has spin and parity 0^+, which is to be expected, and so, if the spin of ThC′ is not 0, the first and third conditions are both taken care of. Now all rotational states of even-even nuclei must have even parity and spins 0,2,4,6...[8] Thus ThC′ may be 0^+ or $2n^+$. If the former, there is no correlation, and the existence of a correlation in an appropriate form, conversely, is evidence that a higher rotational state is involved.

We can narrow the possible I values of ThC′ down even further. ThC is[9] 1^-, and for its decay ΔI can be 0 or ± 1 with a parity change required. It is clear, then, that ThC′ must be either 0^+ or 2^+.

It is necessary to inquire whether the ThC′ state reached by the beta decay is the same as that which emits the alpha particle. The question arises because of the existence of several "long range" alpha groups, corresponding to excited levels in ThC′, which result from lower energy beta groups from ThC. Such levels can also reach the ground state of ThC′ by gamma emission before the alpha decay. Gamma transitions of this kind, however, occur in less than 20 per cent of the beta decays, which means that, regardless of their angular distribution, they should not completely mask the correlation of the principal beta group. Furthermore, the average energy of the minor groups is lower than that of the principal one, so that in any case, the discrimination against low-energy betas required will tend to reduce their effect on the correlation.

In general, the direction-direction correlation between successively emitted nuclear radiations can be given as[6]

$$W(\theta) = \sum_{\nu} A_{\nu} P_{\nu} (\cos\theta) \qquad (2)$$

where $W(\theta)$ is the probability for the angle θ to occur, A_{ν} a constant,

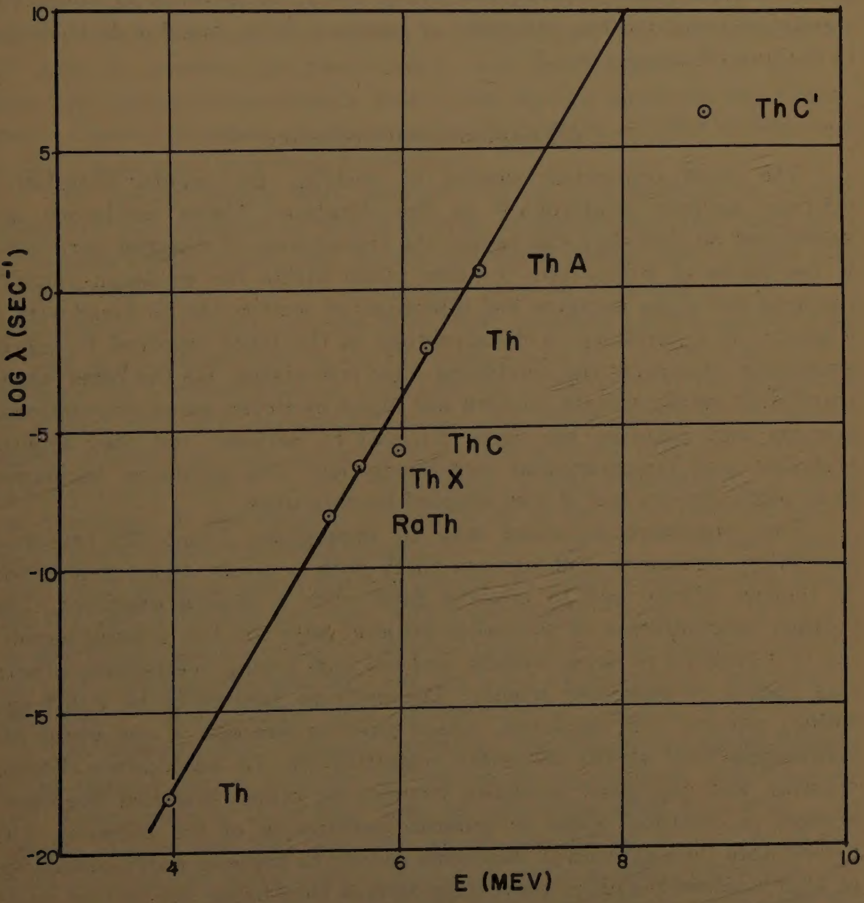

FIGURE 1. The logarithm of the decay constant λ plotted as a function of the alpha particle energy E for the thorium series, with the positions of the various members indicated.

and P_ν the Legendre polynomial of order ν. The complexity of the correlation, *i.e.* the value of ν, is restricted by the condition that $2\nu \leqslant I$, l_1, l_2, where l_1 and l_2 are the angular momenta of the successive radiations. Since the decay of ThC is first-forbidden, ν is either 0 or 2 depending on the spin of ThC′. If $I = 2$, the correlation must be of the form

$$W(\theta) = 1 \pm A \cos^2\theta \qquad (3)$$

The value of A can be calculated theoretically only if the correct electron-neutrino interaction or mixture of interactions is known for the decay of ThC. This information is as yet undetermined. (In fact, beta-correlation theory finds its principal application in finding the nature of the interaction, usually not known *a priori*, from experimental data.) In order to distinguish between $I = 0$ and $I = 2$, however, it is sufficient merely to establish the presence or absence of an angular distribution in the form of EQUATION 3.

(3) *Experimental Procedure*

The most convenient method of studying the angular correlation utilizes nuclear emulsions[10] as the detector. These emulsions are sensitized so that they can record the trajectories of charged particles. If the decay of EQUATION 1 takes place within the emulsion volume, the beta and alpha energies and directions of motion can be found with a minimum of uncertainty. A disadvantage is the labor involved in microscopically examining the emulsions after processing. On the other hand, correlation measurements on beta and alpha particles using conventional sources and counters are quite difficult to perform, and they require elaborate and temperamental instrumentation. The emulsion technique is so much simpler that it was adopted for this work.

The procedure followed was to impregnate Ilford G5 (electron sensitive) emulsions 100 microns thick with a dilute aqueous solution of thorium nitrate and to process them after a week's exposure. The thorium concentration is somewhat critical, with too low a value resulting in a scarcity of decay events, and too high giving overlapping events that cannot be examined readily. The optimum seemed to be 0.004 gm. $ThNO_3$ per cm.^{3} of emulsion, which gave an average of one event per microscope field at 485 diameters magnification. An appropriate thorium solution was deposited in known quantity on plates that had been presoaked in distilled water to promote penetration of the solution. The plates were then allowed to evaporate slowly to dryness under conditions of high relative humidity, preventing strains from being set up that might cause distortion. "Strong" development in amidol[10] was carried out to assure maximum sensitivity, with the attendant disadvantage of a high fog background. In this way, however, even the most energetic electrons produced usable tracks.

The effect of fog background on the detection of low-energy electron tracks has been studied earlier,[11] with the expected result that scanning efficiency decreased with increasing "noise." An unexpected conclusion of that investigation was that "pseudo-tracks," *i.e.* random configurations of background grains interpreted as actual tracks, were identified in numbers comparable with real tracks under conditions of extreme fog. The developed emulsion used here had background of about 0.02 grains/micron,[2] which is the point at which real and pseudotracks had equal probabilities for recognition. As pointed out previously, however, this figure is misleading since it refers to tracks in a horizontal plane. In actual emulsion scanning, the electron track nearly always lies at an angle with the horizontal, requiring the microscope focus to be adjusted in order to observe the whole track, and the possibility for error is therefore reduced considerably below that for the idealized two-dimensional case. The extent of the reduction is perhaps a factor of five, and so the plates used here were satisfactory.

The decay events were searched for at a magnification of 485 diameters and examined at 970 diameters. Measurements were made with an eyepiece micrometer, the microscope depth gauge, and a stage protractor. The latter has the advantage over an eyepiece protractor of much greater accuracy, but the additional accuracy proved unnecessary in view of the uncertainty in the shrinkage factor S. The shrinkage factor is important because what is seen with the microscope is the projection on a plane of a randomly-oriented angle, and, since the emulsion shrinks during processing, the observed track inclinations must be converted to their initial values. In this experiment, S was evaluated by plotting the horizontal X and vertical Z projections of the alpha particle tracks on a graph. Since the track length $R = (X^2 + S^2Z^2)^{1/2}$ is very nearly constant, S is the slope of the line joining the experimental points. S was found to be 2.1 ± 0.2 in this way, with a consequent uncertainty in the calculated angles that depended on the track orientation. The largest combined dip angle encountered was 60°, and the error introduced in this case is less than 5°. Still smaller error figures hold for the rest of the tracks.

The characteristic "stars" formed by the successive alpha decays of the thorium series (FIGURE 2) are readily recognized, and the 8.78 mev alpha particle from ThC′ can be identified from its range. The vicinity of each star was searched for electron tracks originating at the intersection of the alpha tracks, and range, grain density, dip angle, and the projected angle between alpha and beta tracks were ascertained for any such tracks located. Range and dip angle were also measured for the alpha tracks, to permit calculation of the shrinkage factor and the space

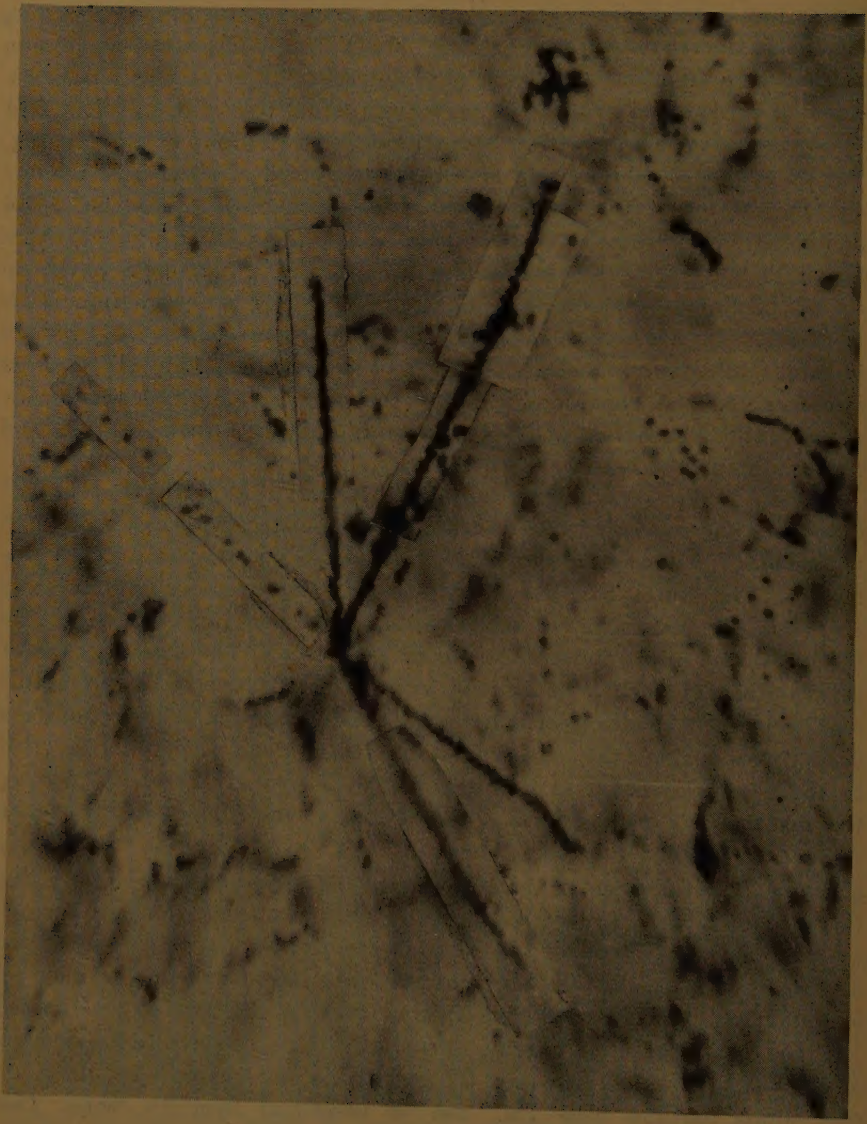

FIGURE 2. A photomicrograph of a typical "star" formed in a nuclear emulsion by the successive decays of ThX, Tn, ThA, ThB, ThC, and ThC′. The beta track from ThC points toward the upper left-hand corner and the alpha track from ThC′ is the long one pointing toward the upper right.

angle between the two tracks. In this work, only entirely unambiguous tracks were measured, which meant that only 119 suitable tracks were found in the approximately 600 events scanned.

(4) *Experimental Results*

For the tracks studied, the grain densities g (in grains/100 microns) and space angles θ were tabulated. The importance of g follows from two considerations. First, any correlation present is to be found only in the higher energy tracks, which exhibit a low grain density. Plotting the angular distribution observed for various intervals of g, then, should show greater asymmetries for the lower g values if any correlation exists. Second, there is some danger of mistaking the electron from the beta-decay of ThB with that from ThC. The average energy of the ThB beta, however, is about 0.1 mev while it is 0.9 mev for ThC, so that, again, the smaller g is, the greater the correlation to be expected.

The experimental results are plotted in FIGURE 3. In view of the relatively small number of events, the angular intervals are 60° wide to give more statistical weight to the distributions. Such a width is permissible since the correlation of EQUATION 3 would still show up if $A \neq 0$.

It is evident from FIGURE 3 that the angular distributions can be fitted by EQUATION 3 using the + sign. The values for the coefficient A corresponding to the different g ranges are given in TABLE 1.

The statistical validity of so small a sample may well be questioned. It is therefore appropriate to compare for the various g ranges the deviation of the number of tracks in the 60° to 120° interval from the mean of the other intervals with the "reliable error" of the mean. The reliable error of a number of counts N is that error for which there is a 90 per cent chance that it will *not* be exceeded, and is given by $1.64\sqrt{N}$. The deviations and the corresponding reliable errors are given in TABLE 2. The deviations exceed the reliable error for $g < 15$, 20, and 30 grains/100 microns, they are about equal for $g < 40$, and the reliable error is significantly greater only for the total sample. Hence there is greater than a 90 per cent probability that a correlation in the form of EQUATION 3 is present for the more energetic electrons. This indicates that, while the assignment of the lowest state of ThC′ as 2^+ cannot be regarded as conclusively established, the present data strongly suggest it.

(5) *Conclusions*

The assignment of 2^+ to the lowest observed level of ThC′ apparently contradicts the empirical rule that the ground states of even-even nuclei are 0^+. ThC′ is highly unstable, and this may account for its anomalous status. Another approach is to consider the 2^+ state as being

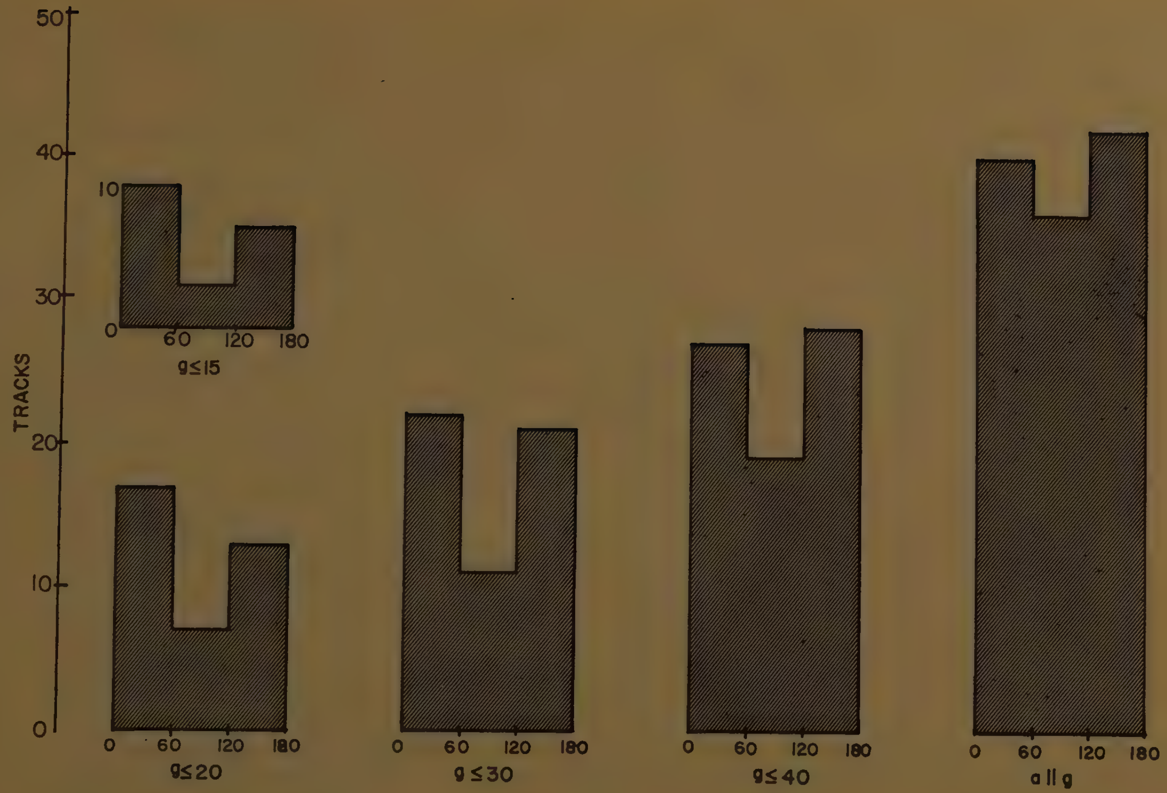

FIGURE 3. The experimental results, plotted as the number of tracks in the 0° to 60°, 60° to 120°, and 120° to 180° angular intervals for various ranges of the grain desity *g*.

TABLE 1

VALUES OF THE COEFFICIENT A IN THE EQUATION $W(\theta) = 1 + A\cos^2\theta$ FOR VARIOUS g RANGES

g	A
$\leqslant 15$	1.8
$\leqslant 20$	1.1
$\leqslant 30$	1.0
$\leqslant 40$	0.45
All	0.13

isomeric, with alpha decay taking place before rotational energy can be lost through other means, *e.g.* quadrupole gamma emission. Such is the case for the higher levels[2] of ThC′, which result in 9.49 and 10.54 mev alpha particle groups. But between 10 and 20 per cent of the 8.78 mev decays occur after the ThC′ nucleus had undergone a transition from one of the higher states, and it seems unrealistic at first glance to suppose that none of these transitions would be to the hypothecated ground state of 0^+. The higher levels of ThC′, however, must have the same spin as the one emitting the 8.78 mev alpha particle, or perhaps a higher value. In either event, their gamma decay to a 0^+ level directly has a very much smaller probability than their decay to the 2^+ level. A number of instances[3] are known where the probabilities for quadrupole emission are small enough so that the decay times exceed 10^{-7} seconds and, if that were true here, it would confirm the supposition that the 2^+ level is actually isomeric. In the absence of both experimental data showing the presence of a less energetic alpha group than the 8.78 mev one (which might be masked by coinciding with one of the other alpha energy groups in the thorium decay scheme or by being very close

TABLE 2

THE DEVIATIONS OF THE NUMBER OF TRACKS IN THE $60°$ TO $120°$ INTERVAL FROM THE MEAN OF THE OTHER INTERVALS AND THE CORRESPONDING RELIABLE ERROR OF THE MEAN FOR THE VARIOUS RANGES OF g

g	Deviation	Reliable Error
$\leqslant 15$	5.5	4.8
$\leqslant 20$	8.0	6.3
$\leqslant 30$	10.5	7.6
$\leqslant 40$	8.5	8.6
All	5.0	10.5

in energy to 8.78 mev, leading in the latter case to an extremely small gamma transition probability) and in the absence also of specific calculations of the quadrupole transition probability here, however, no definite statement can be made.

With regard to the deviation of ThC′ from the Geiger-Nuttall relation, the present investigation indicates that part of the effect may arise from the angular momentum of the decay alpha particle. The ratio between the centrifugal and Coulomb barrier heights for a nucleus of this size is approximately $0.0024l(l+1)$, and, for $l=2$, becomes 0.0144. When this figure is used in calculating the lifetime against alpha decay, the decay constant λ is reduced below that for $l = 0$ by a factor of about 2. This reduction accounts for part of the anomalous ThC′ lifetime. The cause of the remainder of the deviation apparently lies elsewhere.

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